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Specification

1. Title of Invention

Hard coating agent and optical product made of plastic

2. Claims

1. Hard coating agent featured in that it is made up by compounding

(a) epoxy group-containing silicic compound expressed by the following general formula (1), or its partial hydrolyzate



(in the formula, R^1 is an epoxy group-containing organic group of carbon number 2 ~ 8, R^2 is an alkyl group of carbon number 1 ~ 6, alkenyl group, alkyl halide group, or aryl group, R^3 is hydrogen, alkyl group of carbon number 1 ~ 6, alkoxyalkyl group, or acyl group)

and,

(b) tetrafunctional silicic compound expressed by the following general formula (2), or its partial hydrolyzate



(in the formula, R^3 is an epoxy group-containing organic group of carbon number 2 ~ 8)

and,

(c) inorganic fine particles of 1 member or more selected from colloidal titania, colloidal antimony oxide, and colloidal alumina,

and,

(d) curing catalyst.

2. Hard coating agent featured in that it is made up by compounding

(a') partial co-hydrolyzate of epoxy group-containing silicic compound expressed by the following general formula (1)



(in the formula, R^1 is an epoxy group-containing organic group of carbon number 2 ~ 8, R^2 is an alkyl group of carbon number 1 ~ 6, alkenyl group, alkyl halide group, or aryl group, R^3 is hydrogen, alkyl group of carbon number 1 ~ 6, alkoxyalkyl group, or acyl

group) and tetrafunctional silicic compound expressed by the following general formula (2)



(in the formula, R^3 is an epoxy group-containing organic group of carbon number 2 ~ 8) and,

(c) inorganic fine particles of 1 member or more selected from colloidal titania, colloidal antimony oxide, and colloidal alumina,

and,

(d) curing catalyst.

3. Hard coating agent according to the claim 1 or 2 in which the epoxy group-containing silicic compound is 3-glycidoxypolydimethylsiloxane or 2-(3',4'-epoxycyclohexyl)ethylmethyldialkoxysilane, and the tetrafunctional silicic compound is tetramethoxysilane, tetraethoxysilane, methylpolysilicate, or ethylpolysilicate.

4. Hard coating agent according to any of the claims 1 through 3 in which the curing catalyst is aluminum perchlorate.

5. Optical product made of plastic of which the surface is covered with hard coat by the hard coating agent according to any of the claims 1 through 4.

3. Detailed Description of the Invention

Area of application in industry

The present invention relates to the hard coating agent which gives hard coat which is high in hardness, and good in scratch resistance, wear resistance, and further solvent resistance and adhesion, and in addition, excellent in dyeability, and high in refractive index, and also, optical product made of plastic of which the surface is covered with the aforesaid hard coat.

Conventional technology and the problem to be solved by the invention

Nowadays it is a common practice to dye lenses of eyeglasses by disperse dye, etc. in order to give fashionability or antidazzle characteristic. Since this dyeing is carried out after the hard coating processing, it is necessary to cover the eyeglass lenses with highly dyeable hard coat.

Furthermore, recently high refractive index lenses have been used widely in order to make the plastic lens lighter, and for example, the material of the refractive index 1.63 ~ 1.68 has been proposed in JP (Kokai) 64-54021. If hard coat of low refractive index is coated on such high refractive index lenses, interference fringes are generated significantly, and this is not preferable as the optical material, and therefore, it is desirable to coat the hard coating agent which gives high refractive index coat close to the refractive index of the plastic lenses.

On the other hand, in order to give high hardness, wear resistance, solvent resistance, etc. to the optical product made of plastic which is light, and excellent in workability and shock resistance, for example, a coating composition consisting of alkyltrialkoxysilane hydrolyzate and colloidal silica has been proposed (JP 52-39691, JP 62-55554). However, the refractive index of the hard coat given by this composition is low, and in addition, it is not dyeable at all.

Also, in order to prevent corrosion of the metal base, a coating composition, which contains alkylsilanetriol and colloidal silica and colloidal alumina, has been proposed in JP 63-45752. However, although this composition gives coat of a little higher refractive index, interference fringes appear when it is coated on the high refractive index plastic such as polycarbonate resin, thiourethane resin, etc., and in addition, it is not dyeable at all.

Similarly, a composition which contains alkylsilanetriol and colloidal silica and colloidal titania has been proposed in order to improve the weathering resistance of polycarbonate resin, etc. (JP 58-13101). The refractive index of the coat of this composition increases with increasing colloidal titania, but it is not dyeable. Also, a composition which consists of alkylsilanetriol and colloidal silica and colloidal antimony oxide has been proposed in order to give antistatic characteristic. The cured coat of this composition is not quite dyeable either.

In this manner, the coats given by the previous hard coating compositions are in all cases poor in dyeability, and in addition, since their refractive indices are low, they tend to generate interference fringes when the high refractive index lenses are coated, and thus they are not necessarily suitable to coat the optical product made of plastic such as the aforesaid eyeglass lenses, etc..

The present invention has been achieved in the light of the aforesaid situation, and its object is to provide the hard coating agent which gives hard coat which is high in hardness, and good in scratch resistance, wear resistance, and further solvent resistance and adhesion, and in addition, excellent in dyeability, and high in refractive index, and also, optical product made of plastic of which the surface is covered with the aforesaid hard coat.

Method and the function for solving the problems

The present inventors carried out intensive studies on the hard coating agent, and as a result, found that the aforesaid object can be achieved by compounding

(a) epoxy group-containing silicic compound expressed by the following general formula (1), or its partial hydrolyzate



(in the formula, R^1 is an epoxy group-containing organic group of carbon number 2 ~ 8, R^2 is an alkyl group of carbon number 1 ~ 6, alkenyl group, alkyl halide group, or aryl group, R^3 is hydrogen, alkyl group of carbon number 1 ~ 6, alkoxyalkyl group, or acyl group) and,

(b) tetrafunctional silicic compound expressed by the following general formula (2), or its partial hydrolyzate



(in the formula, R^3 is an epoxy group-containing organic group of carbon number 2 ~ 8) or,

(a') co-hydrolyzate of epoxy group-containing silicic compound of the aforesaid formula (1) and tetrafunctional silicic compound of the aforesaid formula (2),

and,

(c) inorganic fine particles of 1 member or more selected from colloidal titania, colloidal antimony oxide, and colloidal alumina,

and,

(d) curing catalyst.

Namely, according to the study by the present inventors, the disperse dye does not penetrate into the cured coat of the hydrolyzate of methyltrialkoxysilane, ethyltrialkoxysilane, etc., and similarly, the disperse dye hardly penetrates into the cured coat of the hydrolyzate of γ -glycidoxypopyltrimethoxysilane, and also, in the co-hydrolyzate of epoxy group-containing bifunctional silane and epoxy group-containing trifunctional silane, it is difficult to make the coat hardness and dyeability compatible, and when the hardness is raised, the dye is hard to penetrate, and when the dye is made penetrating into the coat to some extent, the coat surface tends to be scratched easily. On the other hand, the present inventors found that, by constructing the skeleton of the organopolysiloxane with epoxy group-containing bifunctional silane and tetrafunctional silane, it can be dyed easily with the disperse dye, and thus the dyeability and such characteristics as hardness, scratch resistance, etc. can be made compatible, and in addition, by compounding colloidal titania, colloidal antimony oxide, or colloidal alumina with the aforesaid organopolysiloxane component, the refractive index of the coat, which was as low as 1.45 ~ 1.49 previously, can be raised to 1.55 ~ 1.60 level which is close to the refractive index of the plastic lens, such that the generation of the interference fringes can be prevented, and therefore, it is possible to obtain the hard coating agent which gives hard coat which is high in hardness, and good in scratch resistance, wear resistance, and further solvent resistance and adhesion, and in addition, excellent in dyeability, and high in refractive index, and also, suitable for optical product made of plastic, and completed the present invention.

Consequently, the present invention provides the hard coating agent featured in that it is made up by compounding

(a) the epoxy group-containing silicic compound of the aforesaid formula (1) or its partial hydrolyzate, and

(b) the tetrafunctional silicic compound of the aforesaid formula (2) or its partial hydrolyzate, and

(c) inorganic fine particles of 1 member or more selected from colloidal titania, colloidal antimony oxide, and colloidal alumina, and

(d) curing catalyst,

and the hard coating agent featured in that it is made up by compounding

(a') co-hydrolyzate of epoxy group-containing silicic compound of the aforesaid formula (1) and tetrafunctional silicic compound of the aforesaid formula (2),

and,

(c) inorganic fine particles of 1 member or more selected from colloidal titania, colloidal antimony oxide, and colloidal alumina,

and,

(d) curing catalyst,

and the optical product made of plastic of which the surface is covered with the hard coat by the aforesaid hard coating agent.

In the following, the present invention is explained in detail further.

The (a) constituent of the hard coating agent of the present invention contributes to the coat formation and adsorption of organic dye, and it is epoxy group-containing silicic compound expressed by the following general formula (1) or its hydrolyzate.



The substituent R^1 in the formula here is an epoxy group-containing organic group of carbon number 2 ~ 8, and specifically it can be a glycidoxyalkyl group which has alkylene group of carbon number 1 ~ 4 such as glycidoxymethyl group, glycidoxypropyl group, etc., or epoxycyclohexylalkyl group, and so forth. Also, R^2 is an alkyl group of carbon number 1 ~ 6, alkenyl group, alkyl halide group, or aryl group, and in this case, the alkyl halide group is preferably of carbon number 1 ~ 6, the alkenyl group is of carbon number 2 ~ 6, and the aryl group is of carbon number 6 ~ 10, and specifically, they can be methyl group, ethyl group, propyl group, 3-chloropropyl group, 3,3,3-trifluoropropyl group, vinyl group, allyl group, phenyl group, etc.. R^3 is a hydrogen, alkyl group of carbon number 1 ~ 6, alkoxyalkyl group, or acyl group, and among them, the alkyl group is preferably of carbon number 1 ~ 4, the alkoxyalkyl group is of carbon number 2 ~ 8, and particularly 3 ~ 8, containing alkoxy group of carbon number 1 ~ 4, and the acyl group is of carbon number 2 ~ 6, and specifically, they can be hydrogen atom, methyl group, ethyl group, propyl group, butyl group, isopropyl group, isobutyl group, methoxyethyl group, ethoxyethyl group, butoxyethyl group, acetyl group, propionyl group, etc..

Such silicic compound of the formula (1) can be, for example, 3-glycidoxypropylmethyldimethoxysilane, 3-glycidoxymethyldiethoxysilane, 3-glycidoxypropylphenyldimethoxysilane, 2-(3',4'-epoxycyclohexyl)ethylmethyldimethoxysilane, 2-(3',4'-epoxycyclohexyl)ethylmethyldiethoxysilane, etc., and they can be used individually or as a combination of two or more. Incidentally, in the present invention 3-glycidoxypropylmethyldialkoxysilane and 2-(3',4'-epoxycyclohexyl)ethylmethyldialkoxysilane are particularly suitable.

Furthermore, although the epoxy group-containing silicic compound of the formula (1) can be used as is, it also can be used as the partial hydrolyzate by partially hydrolyzing it in advance.

Incidentally, it is usually preferable that the aforesaid epoxy group-containing silicic compound is used by dissolving in organic solvent such as alcohol, ketone, ester, cellosolve, aromatic compound, etc..

Now, in the present invention, the tetrafunctional silicic compound as the (b) constituent contributes to the strength of the coat, adhesion of the coat to the plastic base, and also to fixing of the (c) constituent described later, and it is the tetrafunctional silicic compound expressed by the following general formula (2) or its partial hydrolyzate.



(R^3 in the formula is the same as R^3 in the aforesaid formula (1)).

Specifically, the compound of the formula (2) can be tetramethoxysilane, tetraethoxysilane, tetrabutoxysilane, methylpolysilicate, ethylpolysilicate, etc., and they can be used individually or as a combination of two or more. In the present invention, tetramethoxysilane, tetraethoxysilane, methylpolysilicate, and ethylpolysilicate are used particularly suitably.

Furthermore, the compounding amount of the (b) constituent should be 2 ~ 200 parts (by weight, same hereafter), preferably 5 ~ 100 parts, against 100 parts of the (a) constituent, and if the compounding amount is less than 2 parts, the coat hardness may be

lower, and if it exceeds 200 parts, the coat may become fragile and its covering power may be degraded.

In the present invention, the aforesaid (a) constituent and the (b) constituent can be replaced by the (a') constituent, which is the co-hydrolyzate of the epoxy group-containing silicic compound of the aforesaid formula (1) and the tetrafunctional silicic compound of the aforesaid formula (2), namely, the organopolysiloxane obtained by co-hydrolyzing the compound of the formula (1) and the compound of the formula (2) in advance can be compounded.

Incidentally, in this case, the co-hydrolysis can be carried out by the common method, and also the compounding amount of the (b) constituent should be the same as the foregoing.

Also, in the hard coating agent of the present invention, in addition to the aforesaid silicic compounds which are the essential constituents, other silicic compound, for example, methyltrialkoxysilane, phenyltrialkoxysilane, vinyltrialkoxysilane, dimethyldialkoxysilane, diphenyldialkoxysilane, phenylmethyldialkoxysilane, vinylmethyldialkoxysilane, 3-methacryloxypropyltrialkoxysilane, 3-methacryloxypropylmethyldialkoxysilane, etc. can be added as the arbitrary constituent. Incidentally, although there is no particular limitation for the compounding amount for the aforesaid other silicic compound, it is preferable not to exceed 100 parts against the (a) constituent or the (a') constituent 100 parts for the sake of their dyeability after curing.

Now, the inorganic fine particles, the (c) constituent of the hard coating agent of the present invention, contributes to the coating agent by increasing the coat hardness and the refractive index of the coat, and 1 member or more selected from colloidal titania, colloidal antimony oxide, and colloidal alumina are compounded.

Here, these inorganic fine particles are preferably of the particle size 1 ~ 200 μm , and particularly 5 ~ 100 μm , and if the particle size is less than 1 μm , the surface hardness of the coat may be degraded, and if it exceeds 200 μm , the transparency of the coat may be degraded.

Also, such inorganic fine particles used can be those colloid solution on the market in which inorganic fine particles are dispersed in water or organic solvent, and for example, they can be Alumina Sol 520, San Colloid NTS-10 (both are products of Nissan Chemical), Alumina Clear Sol (product of Kawaken Fine Chemical), etc..

Furthermore, since the refractive index of the coat can be controlled by the compounding amount of the (c) constituent, it is properly selected depending on the desired refractive index of the coat, but it is preferably 10 ~ 300 parts, and particularly 20 ~ 200 parts as solid of the inorganic fine particles against the (a) constituent 100 parts. If the compounding amount of the (c) constituent is less than 10 parts, the coat of high hardness cannot be obtained and also the refractive index of the coat may be low, and if it exceeds 300 parts, the coat obtained is brittle, and its covering power may be poor.

The (d) constituent of the present invention is curing catalyst which is needed to cure the mixture of the aforesaid essential constituents.

In this case, in consideration of stability of the coating agent, hardness of the coat, non-yellowing characteristic, curing characteristic, etc., for example, Lewis acid catalyst, organoaluminum compound, organotitanium compound, etc. are used suitably as the curing catalyst, and specifically, it can be aluminum chloride, aluminum perchlorate,

aluminum phosphate, aluminum triisopropoxide, aluminum acetylacetonate, tetrabutyl titanate, tetraisopropyl titanate, etc., and among them, aluminum perchlorate is used preferably.

Furthermore, the amount of addition of the (d) constituent can be 0.05 ~ 10 parts, and particularly 0.1 ~ 5 parts, against the (a) constituent 100 parts, and if the amount of its addition is less than 0.05 part, the curing characteristic of the coating agent may be degraded, and if it exceeds 10 parts, the stability of the coating agent may be degraded.

In order to improve the adhesion to the plastic base, weather resistance, workability of coating, antireflection characteristic, etc. of the hard coating agent of the present invention, various constituents can be added as long as the effect of the present invention is not impeded. For example, epoxy resin such as polyolefin base epoxy resin, cyclohexenoxide polyglycidylester, condensation polymer of epichlorohydrin and bisphenol A, copolymer with glycidylmethacrylate and acrylic compound, etc., and ultraviolet absorbent of benzophenone base, benzotriazole base, phenol base, etc. can be added. Furthermore, in order to improve the coating characteristic, various surfactant can be compounded, and in particular, block copolymer or graft copolymer of dimethylsiloxane and polyether, fluorine base surfactant, etc. are effective.

The coating agent of the present invention can be obtained by adding the (c) constituent and the (d) constituent after mixing the (a) constituent and the (b) constituent, or the (a') constituent, and arbitrary constituents, as needed, and in particular, when the organosilicic compound and its partial hydrolyzate are used, it can be obtained by mixing the (a) constituent and the (b) constituent, and arbitrary constituents as needed, and then adding water or acidic solution such as hydrochloric acid, acetic acid, etc. for hydrolysis, and then adding the (c) and (d) constituents to this. Also, in case the (c) constituent is acidic solution or basic solution, it is possible to carry out the hydrolysis with the acidic substance or basic substance contained in the (c) constituent by mixing the (a) constituent, the (b) constituent, and the (c) constituent.

Incidentally, the solvent which can be contained in the coating agent of the present invention is preferably lower alcohol such as methanol, ethanol, isopropanol, butanol, isobutanol, diacetonealcohol, etc., or cellosolve such as methyl cellosolve, ethyl cellosolve, cellosolve acetate, butyl cellosolve, etc., and its mixture with such solvent as ester, ketone, amide, aromatic compound, etc. can be also used suitably.

The hard coating agent of the present invention can be applied on various plastics. There is no particular limitation for the plastic material on which the hard coating agent of the present invention can be applied, and they can be polycarbonate, polystyrene, polyester, polyurethane, or polythiourethane. The hard coating agent of the present invention is coated on the optical product made of plastic such as plastic lenses, and by the heat treatment, protective coat of high hardness can be obtained. The coating method can be a common coating method such as brush coating, roll coating, spray coating, flow coating, dip coating, spin coating, etc.. Further, the curing condition varies by the compounding amount of the curing catalyst, etc., but the cured coat of the object can be obtained by curing at the temperature lower than the softening point of the plastic base, usually 80 ~ 150°C, for 0.5 ~ 10 hours. Also, the cured coat obtained by the coating agent of the present invention can be dyed, and the dyeing condition is determined arbitrarily for the concentration of the various disperse dyes, temperature, time, etc..

Usually, it can be easily dyed by soaking in the dye bath, in which 0.1 ~ 1 weight % dye is dispersed, for 5 ~ 15 minutes at 80 ~ 100°C.

Effect of the invention

As explained in the foregoing, the hard coating agent of the present invention gives hard coat which is of high hardness, scratch resistant, wear resistant, and further, excellent in solvent resistance, adhesion, etc., and in addition, excellent in dyeability and can be easily dyed by disperse dye, etc., and also is of high refractive index so that it does not generate interference fringes even if it is coated on the plastic base with the refractive index higher than 1.5. Consequently, it is possible to obtain optical product made of plastic which is highly transparent and of good appearance by forming hard coat on the surface by the hard coating agent of the present invention.

<Examples, comparison examples>

In the following, the present invention is explained specifically by examples and comparison examples, but the present invention is by no means limited by the following examples.

[Example 1]

3-glycidoxypopylmethyldiethoxysilane 35 g and tetramethoxysilane 5 g were poured into a flask, and while they were being stirred under ice cooling, 0.05 normal hydrochloric acid solution 7.5 g was dropped over 30 minutes. Further, antimony pentaoxide sol (methanol solution, nonvolatile component 30 %) 200 g was added, and digested for 16 hours at 20 ~ 25°C, and then ethanol 20 g, diacetonealcohol 80 g, and aluminum perchlorate hexahydrate 0.6 g were added, and thus the coating liquid was prepared. Subsequently, this coating liquid was coated on alkali-treated CR-39 (diethyleneglycolbisallylcarbonate) by dipping method, and cured at 120°C for 60 minutes.

Also, the aforesaid coating liquid was coated on an extruded polycarbonate plate of the refractive index 1.58 by dipping method, and cured at 120°C for 60 minutes, and inspected interference fringes by visual observation.

[Example 2]

Titania sol (aqueous solution, nonvolatile component 10 %) 200 g was added instead of antimony pentaoxide in the example 1, and digested similarly to the example 1, and then ethanol 10 g, diacetonealcohol 20 g, and aluminum perchlorate hexahydrate 0.6 g were added, and thus the coating liquid was prepared, and similarly to the example 1, it was coated on the CR-39 and polycarbonate plate, and cured.

[Example 3]

Alumina sol (aqueous solution, nonvolatile component 10 %) 200 g was added instead of antimony pentaoxide in the example 1, and digested similarly to the example 1, and then ethanol 10 g, diacetonealcohol 20 g, and aluminum chloride hexahydrate 1.2 g were added, and thus the coating liquid was prepared, and similarly to the example 1, it was coated on the CR-39 and polycarbonate plate, and cured.

[Comparison example 1]

3-glycidoxypopyltrimethoxysilane 40 g and isobutyl alcohol 15 g were poured into a flask, and while they were being stirred under ice cooling, 0.05 normal hydrochloric acid solution 9 g was dropped over 30 minutes. Further, antimony pentaoxide sol (methanol solution, nonvolatile component 30 %) 200 g was added, and digested for 16 hours at 20 ~ 25°C, and then ethanol 20 g, diacetonealcohol 80 g, and aluminum

perchlorate hexahydrate 0.6 g were added, and thus the coating liquid was prepared. Subsequently, this coating liquid was coated similarly to the example 1 on the CR-39 and polycarbonate plate, and cured.

[Comparison example 2]

Coating liquid was prepared by the same manner as the example 1 except that silica sol (methanol solution, nonvolatile component 30 %) was used instead of antimony pentaoxide sol in the example 1, and coated on the CR-39 and polycarbonate plate, and cured.

The characteristics of the coat obtained by the aforesaid examples 1 ~ 3 and comparison examples 1, 2 were studied by the following methods. The results are shown in Table 1.

Wear resistance

The coat was rubbed 10 times with #0000 steel wool with 500 g load, and the condition of scratch was evaluated by visual observation.

Adhesion

Following JIS K-5400 paragraph 6.15, the coat was cut to square pattern by 11 lines with 1 mm space in the x and y directions using a knife, and after sticking cellophane adhesive tape (product of Nichiban Co.), a peeling test was carried out, and the number of squares which were not peeled was studied.

Solvent resistance

The coat was rubbed lightly with acetone-impregnated cotton ball 100 times, and the transparency was evaluated visually.

Discoloration

The coat after curing was evaluated visually.

Dyeability

The coated CR-39 lens was soaked in 0.2 weight % aqueous solution of the disperse dye Brown D (product of Seiko Co.) at 87°C for 5 minutes, and the light transmittance of the lens was measured.

Refractive index

After the coating agent was coated on polytetrafluoroethylene sheet and cured at 120°C for 60 minutes, the coat obtained was peeled, and this cured film was sandwiched between methyl salicylate, and using the refractometer (Abbe refractometer, product of Atago Co.), the value before or after the refractive index of methyl salicylate at 25°C (1.5350 at 25°C) was measured.

Interference fringes

The coating liquid was coated on the polycarbonate plate (refractive index 1.587) as described in the foregoing, and the appearance was evaluated visually.

Table 1

	Example			Comparison example	
	1	2	3	1	2
Wear resistance	No change	←	←	←	←
Adhesion	100/100	←	←	←	←
Solvent resistance	No change	←	←	←	←
Dyeability (%)	53	45	40	84	55
Refractive index	1.582	1.580	1.522	1.581	1.490
Interference fringes	None	None	None	None	Many

From the results in Table 1, it was found that the hard coat by the hard coating agent of the present invention is good in wear resistance, solvent resistance, and adhesion, and in addition, excellent in dyeability, and further, the refractive index is as high as over 1.5, and no interference fringe is observed when it is coated on the extruded polycarbonate plate.

Applicant
Representative

Shin-Etsu Chemical Co., Ltd.
Takashi Kojima (and another)